22: Congruent Melting Kinetics: Constraints on Chondrule Formation

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ABSTRACT

The processes and mechanisms of melting and their applications to chondrule formation are discussed. A model for the kinetics of congruent melting is developed and used to place constraints on the duration and maximum temperature experienced by the interiors of relict-bearing chondrules. Specifically, chondrules containing relict forsteritic olivine or enstatitic pyroxene cannot have been heated in excess of 1901°C or 1577°C, respectively, for more than a few seconds.

INTRODUCTION

Since the discovery of relict grains by Nagahara (1981) and Rambaldi (1981), the emerging models of chondrule formation have called for an origin by the melting of pre-existing solids (e.g. Grossman, 1988). The realization that the majority of FeO-rich chondrules have lost little Na (Grossman, 1988; Hewins, 1991a; Grossman, this volume), coupled with experimental work on Na loss from chondritic melts (Tsuchiyama *et al.*, 1981) has led to the idea that chondrules were melted in flash heating events of an unknown dynamical nature (Grossman, 1988; Boss, this volume, Concise Guide).

If chondrules were formed by the melting and dissolution of minerals, then a better understanding of these two processes is needed, in order to derive constraints on the nature of the flash heating event(s). Congruent melting is the process by which a solid transforms above its melting point to a liquid of the same composition, and will be the focus of this paper. Incongruent melting occurs when a mineral solid-solution forms a liquid of different composition when heated above the solidus but below the liquidus. In a study of the incongruent melting of plagioclase (Tsuchiyama and Takahashi, 1983) it was found that the kinetics of the reaction were rate-limited by solid-state diffusion, as the solid also needs to change composition, in order to maintain equilibrium. The sluggish kinetics of this type of process suggest that it will be unimportant during flash heating. Above the liquidus, a mineral solid solution such as plagioclase or olivine will melt congruently (Greenwood and Hess, 1995), as neither the liquid nor the solid must change composition during the transition. Dissolution is the process by which a mineral dissolves into a liquid of different composition, and this occurs below the melting point of the solid. Dissolution should be an important process during chondrule formation (Greenwood and Hess, 1995), but will not be discussed here.

In this paper, the mechanisms and kinetics of congruent melting are examined. It is shown that the kinetics of congruently melting minerals are best described by an interface-controlled model (Wilson, 1900; Frenkel, 1932). This model is used to calculate the melting rates of possible precursor minerals (Hewins, 1991b), which leads to constraints on the durations and peak temperatures of chondrule formation. The implications of congruent melting during chondrule formation are considered below.

CONGRUENT MELTING: THEORY

Melting as a Continuous Transition

Studies of the melting transition in congruently melting materials have historically focused almost exclusively on the instability of the solid while neglecting the parallel reaction, the growth of the liquid (Boyer, 1985). At the melting point, homogeneous melting models envision the solid catastrophically transforming to liquid at all points in the crystalline lattice. Nucleation of the liquid is not necessary. Various theories have been developed to explain this bulk mechanical instability. Some examples of these are the Lindemann criterion which links the melting point to a critical amplitude of atomic vibrations (Lindemann, 1910), the vanishing of the shear modulus (Born, 1939), and the generation of dislocations (Poirier, 1986).

Continuous melting models are necessary, but insufficient components to our understanding of the melting transition. Phenomena

such as superheating (exposing the solid to temperatures above T_m, the melting point) (Di Tolla *et al.*, 1995) and surface melting (melting preferentially at surfaces of the crystal) (Frenken and van Pinxteren, 1994) are not predicted by these theories. Though it has long been known that some silicates can sustain large amounts of superheat for considerable lengths of time (Day and Allen, 1905), the proponents of continuous melting models were driven by the early experimental observations that metals were melted almost instantaneously at fractions of a degree of superheat (Ainslie *et al.*, 1961). In fact, metals can be superheated by several degrees (Dages *et al.*, 1987; Di Tolla *et al.*, 1995), but due to the high rate of melting it is generally difficult to observe. Also, homogeneous melting models are at odds with the observation that melting is invariably initiated at external surfaces (Tamman, 1925; Teraoka, 1993) and internal cracks and cleavage planes (Uhlmann, 1980). The importance of the surface in initiating melting during molecular dynamics simulations of $MgSiO_3$-perovskite has also been discussed by Belonoshko (1994).

Melting as a Discontinuous Transition

The contrasting view of melting is that the solid transforms to a liquid discontinuously via a nucleation and growth mechanism, similar to crystallization (Tamman, 1925). A heterogeneous model would predict that melt will form where the barrier to nucleation of the melt phase is lowest, such as surfaces and lattice defects (Ainslie *et al.*, 1961). This agrees well with experimental observations.

If melting is considered analogous to crystallization, the principal difference being that liquids nucleate far more easily than crystals (Ainslie *et al.*, 1961), then growth of the melt phase can be modelled with existing theories of crystal growth. This approach has been utilized previously in the melting of silicates and oxides (e.g. Wagstaff, 1969; Uhlmann, 1971), and is described below.

CONGRUENT MELTING

Wilson-Frenkel Model

When a solid melts to a liquid of the same composition, it is found experimentally that the growth of the melt is inversely proportional to the viscosity of the liquid phase (Ainslie *et al.*, 1961). In melts with low viscosity, such as metals and semiconductors, growth of the melt is very fast and is generally rate limited by how fast heat can be added to the interface (Spaepen and Turnbull, 1982). In melts with high viscosity, such as silicates, growth is relatively slow and is usually rate limited by the kinetics of the solid-liquid transition. Several studies of the melting kinetics of silicates and oxides have been completed to date (e.g. quartz, Scherer *et al.*, 1970; sodium disilicate, Fang and Uhlmann, 1984; diopside, Kuo and Kirkpatrick, 1985; albite, Greenwood and Hess, 1994a). Each used a Wilson-Frenkel model for normal growth to model their data. A normal growth model is used when the interface is rough on the atomic scale (atoms can be added or removed from any site on the interface). The growth of the melt is envisioned as the propagation of the solid-liquid interface from the surface into the crystal. Molecular dynamics simulations of the melting of forsterite have found that melting takes place layer-by-layer as the solid-liquid interface migrates through the crystal (Kubicki and Lasaga, 1992), in accordance with the tenets of an interface-controlled growth model. The normal growth model has been found to reproduce experimentally determined melting rates generally within an order of magnitude (Greenwood and Hess, 1994b; Table 22.1). The Wilson-Frenkel model is (Uhlmann, 1971):

$$u = (fD'/a_o)[1-\exp(-\Delta G/kT)] \quad (1)$$

where u is the growth rate, f is the fraction of sites available (f=1 for melting; i.e. atoms leaving the crystal are not limited to rigid, fixed sites in the melt), D′ is the kinetic factor for transport at the interface, a_o is the jump distance (in this model, it is usually taken as twice the length of an important bond in the crystal structure; e.g. 2 × Si-O for quartz, Ainslie *et al.*, (1961)), T is the absolute temperature, ΔG is the free energy change per atom of the transition at the temperature T, and k is Boltzmann's constant. If D′ is related to self-diffusion in the liquid, D, and the Stokes-Einstein relation for diffusion is assumed (see discussion below), then:

$$D' \approx D = kT/(3\pi a_o \eta) \quad (2)$$

where η is the viscosity of the melt, and substituting in (1),

$$u = [fkT/(3\pi a_o^2 \eta)][1-\exp(-\Delta G/kT)] \quad (3).$$

If ΔG<<kT, a condition satisfied for small superheats, we expand (3) (Fine,1964) to

$$u = [fkT/(3\pi a_o^2 \eta)](-\Delta G/kT) \quad (4).$$

Also, at small departures from equilibrium, $\Delta G \approx \Delta H_f \Delta T/T_m$ (Kingery *et al.*, 1976), and substituting into (4):

$$u = f\Delta H_f \Delta T/(T_m 3\pi a_o^2 \eta N_A) \quad (5)$$

where ΔH_f is the molar heat of fusion, N_A is Avogadro's number, T_m is the melting point, and $\Delta T = T - T_m$, the amount of superheat. Notice that we are now describing the melting per mole rather than per atom. The functional form of the equation is also written as:

$$u\eta = K\Delta T \quad (6)$$

where $K = f\Delta H_f/(T_m 3\pi a_o^2 N_A)$, and is considered a constant for small superheatings. The parameter uη is termed the "normalized melting rate", and a plot of uη versus ΔT should be linear with a slope equal to K for normal growth (Fang and Uhlmann, 1984). Any deviations from a linear plot would suggest a significant temperature dependence of f, ΔH_f, or a_o (Wagstaff, 1969). An experimentally determined plot of uη versus ΔT for albite is shown in Fig. 1 (Greenwood and Hess, 1994a; Greenwood and Hess, unpublished data) and demonstrates the validity of using a normal growth model to describe the melting of albite. The normal growth model has also been shown to be appropriate for quartz (Scherer *et al.*, 1970), cristobalite (Wagstaff, 1969), sodium disilicate (Fang and Uhlmann, 1984), germanium dioxide (Vergano and Uhlmann, 1970), phosphorus pentoxide (Cormia *et al.*, 1963a), and diopside (Kuo and Kirkpatrick, 1985).

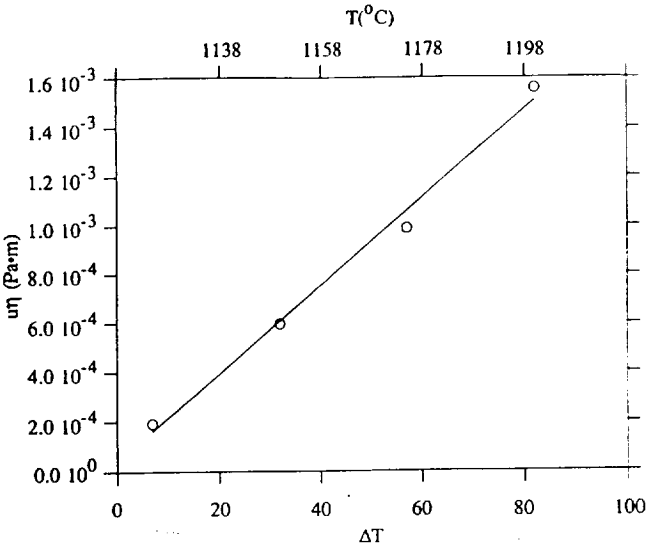

Fig. 1. Shown here is the experimentally determined normalized melting rate, uη (melting rate × viscosity) vs. ΔT (the amount of superheat) for albite. Line is a best fit to the data (open circles).

Comparison of experimental and calculated rates

A comparison of rates calculated from equation (5) with experimentally determined rates is shown in Table 1. A jump distance of 3Å was used for the calculations, except for albite and germanium dioxide, where a value of 3.5Å was used. As mentioned above, the jump distance is approximated as twice the length of an important bond in the structure.

While there is some obvious disagreement, the calculated rates are generally the same order of magnitude as the rates determined by experiment. The largest discrepancies between the calculated and experimental rates are for quartz and diopside, and illustrate some of the experimental difficulties associated with measuring melting kinetics. The melting kinetics of quartz, as well as the viscosity of liquid SiO_2, are highly sensitive to atmospheric impurities and water contamination, and large differences in melting rates have been found by different researchers for quartz (Ainslie *et al.*, 1961; Scherer *et al.*, 1970). The reason for the large discrepancy for diopside is not known, but may be related to the difficulty in measuring high melting rates in a small range of superheating. A similar problem was found in the determination of the melting rates of sodium disilicate, where a second study by the same research group found very different results (Meiling and Uhlmann, 1967; Fang and Uhlmann, 1984). At these high rates of melting, a heating stage may be necessary for accurate determination of the kinetics (Fang and Uhlmann, 1984).

The Wilson-Frenkel model reproduces the experimental data in five of the seven studies shown in Table 1 within an order of magnitude. While experimental difficulties may explain some of the

Table 1. *Comparison of experimental melting rates with rates calculated from eqn (5) at various superheats*

Mineral	ΔHf(kJ/mol)	Tm(°C)	T(°C)	η(Pa•s)	ΔT	Expt./Calc.
Quartz[†1]	9.40[2]	1427[2]	1500	9.71×10^{7} [3]	73	20.5
			1600	1.50×10^{7}	173	24.0
			1650	6.26×10^{6}	223	10.4
Cristobalite[4]	8.92[2]	1726[2]	1743	$1,39 \times 10^{6}$ [3]	17	6.3
			1746	1.31×10^{6}	20	7.9
			1755	1.15×10^{6}	29	9.7
$Na_2Si_2O_5$[5]	37.7[2]	874[2]	875	881[5]	1	2.1
			880	805	6	2.1
			884	662	10	1.8
GeO_2[6]	15[6]	1114[6]	1119	2.95×10^{4} [7]	5	3.6
			1125.5	2.69×10^{4} [7]	11.5	2.5
			1130	5×10^{4} [8]	22	5.1
Albite[9]	64.5[2]	1118[9]	1125	4.3×10^{6} [10]	7	0.4
			1175	1.1×10^{6}	57	0.4
			1200	5.9×10^{5}	82	0.3
P_2O_5[11]	21.8[11]	580[11]	589	4.38×10^{5} [12]	9	0.5
			593	3.61×10^{5}	16	0.9
			609	2.53×10^{5}	29	0.8
Diopside[13]	137.7[2]	1391[13]	1393	0.93[14]	2	0.013
			1399	0.91	8	0.013
			1412	0.87	21	0.012

[†]Metastable melting, [1]Scherer *et al.* (1970), [2]Richet and Bottinga (1986), [3]Urbain *et al.* (1982), [4]Wagstaff (1969), [5]Meiling and Uhlmann (1967), [6]Vergano and Uhlmann (1970), [7]Fontana and Plummer (1966), [8]Sharma *et al.* (1979), [9]Greenwood and Hess (1994a), [10]Stein and Spera (1993), [11]Cormia *et al.* (1963a), [12]Cormia *et al.* (1963b), [13]Kuo and Kirkpatrick (1985), [14]calculated from Bottinga and Weill (1972).

discrepancy, the equation may be fundamentally flawed, and this is considered in the next section.

Discussion of Wilson-Frenkel model

The assumptions used to derive eqn. (2) are somewhat controversial. The first assumption is that the diffusion in the interfacial region can be equated to self-diffusion in the liquid. Cahn *et al.* (1964) suggest that diffusivity in the interfacial region may be as much as two orders of magnitude lower than diffusion in the bulk liquid. In their theory, lower diffusivities arise from the quasi-crystalline nature of the liquid immediately adjacent to the interface. In contrast, molecular dynamics simulations of argon suggest that the diffusivity at the interface may be higher than in the bulk liquid (Broughton *et al.*, 1982). The nature of the solid-liquid interface is not fully understood and is the subject of ongoing work (e.g. Moss and Harrowell, 1994).

The second assumption used in eqn. (2) is the application of the Stokes-Einstein equation to relate self-diffusion in the liquid and the viscosity of the liquid. There has been some success in using the Stokes-Einstein equation to model the diffusion of oxygen (Shimuzu and Kushiro, 1984) and silica (Watson and Baker, 1991) in silicate melts, but in general the quantitative agreement is poor (Kress and Ghiorso, 1995). Although the assumptions used in eqn. (2) may be the source of divergence from the experimental rates, the good agreement for the calculated rates of albite and sodium disilicate suggests that the Stokes-Einstein equation may be appropriate for the melting of silicates. The viscosities of molten albite and sodium disilicate at their melting points are 10^6 and 10^2 Pa•s, respectively (Table 1). They also have different melt structures, a fully polymerized albite melt and a somewhat depolymerized disilicate melt. Considering the large differences in viscosity, melt structure, and melting rates between these two materials, the agreement between the experimental and calculated rates harbors hope for the possibility of using the Wilson-Frenkel model as a predictive vehicle. We feel that this model can be used to predict melting rates within an order of magnitude, and as will be shown below, even two orders of magnitude difference will still lead to useful constraints on chondrule formation.

MELTING DURING CHONDRULE FORMATION

In this section we consider the melting kinetics of possible chondrule precursor minerals (Hewins, 1991b) and relict grains (Jones, this volume). As the relict grains are, by definition, the only precursor minerals to survive the last heating event, constraints on possible precursors and their grain sizes are poor. A mean chondrule diameter of ~1 mm (Grossman *et al.*, 1988) is taken as a maximum precursor grain size, though some chondrules are undoubtedly larger. A minimum grain size for precursor minerals cannot be quantified, but it is noted that if the grain size is very small (~100 Å), the melting points may be lowered due to size-dependent melting (Allen *et al.*, 1986). The presence of fine-grained rims and matrix (Alexander *et al.*, 1989) suggests that material in the micron to submicron range was present in the chondrule forming region. In the analysis below we consider grains in the range of 1000 μm to 10 μm.

Precursor minerals

The melting rates of possible chondrule precursor minerals calculated from eqn. (5) are listed in Table 2 for superheats of 5 and 100 degrees. The jump distance is taken as 3Å (except albite; 3.5Å). Values for the viscosities of fayalite, diopside, åkermanite, and enstatite melts were determined from Bottinga and Weill (1972). For forsterite melts, viscosities were first determined by calculating values in the 1600 –1800°C range by the method of Bottinga and Weill 1972), and then extrapolating to the desired temperature. The viscosities for albite and anorthite melts were determined experimentally (Stein and Spera, 1993; Cranmer and Uhlmann, 1981).

Shown in Fig. 2 are the melting rates of possible precursor minerals at 1600°C. If the melting rates for these minerals are the right order of magnitude, it becomes apparent that if any of these possible chondrule precursors are exposed to temperatures in excess of 1600°C (with the exception of forsterite, T_m = 1901°C) they will probably melt in seconds or less. In fact, for a grain radius range of 10 –100 μm, even two orders of magnitude error for the calculated melting rates still leads to complete melting of chondrule precursor minerals in seconds or less. At higher temperatures, the melting

Table 2. *Melting rates calculated from eqn. (5) at superheast of 5 and 100 degrees*

Mineral	Tm(°C)	Hf (KJ/mol)	ΔT	T(°C)	Calc. rate (μm/s)	ΔT	T(°C)	Calc. rate (μm/s)
Albite*	1118[1]	64.5[2]	5	1123	4.3×10^{-5}	100	1218	4.3×10^{-3}
Fayalite	1217[2]	89.3[2]	5	1222	5×10^{3}	100	1317	1.7×10^{5}
Diopside*	1391[3]	137.7[2]	5	1396	11	100	1492	3.8×10^{2}
Åkermt.	1458[4]	123.6[4]	5	1463	1.6×10^{3}	100	1558	1.3×10^{5}
Anorthite	1557[2]	133.0[2]	5	1562	1.9×102	100	1657	8.2×10^{3}
Enstatite†	1557[5]	73.2[2]	5	1562	n.a.†	100	1657	2.7×10^{4}
Forsterite	1901[6]	142[6]	5	1906	6.3×10^{3}	100	2001	1.3×10^{5}

*Experimentally determined (see text), †See text for discussion of enstatite melting, [1]Greenwood and Hess (1994a), [2]Richet and Bottinga (1986), [3]Kuo and Kirkpatrick (1985), [4]Hemingway *et al.* (1986), [5]Bowen and Anderson (1914), [6]Richet *et al.* (1993).

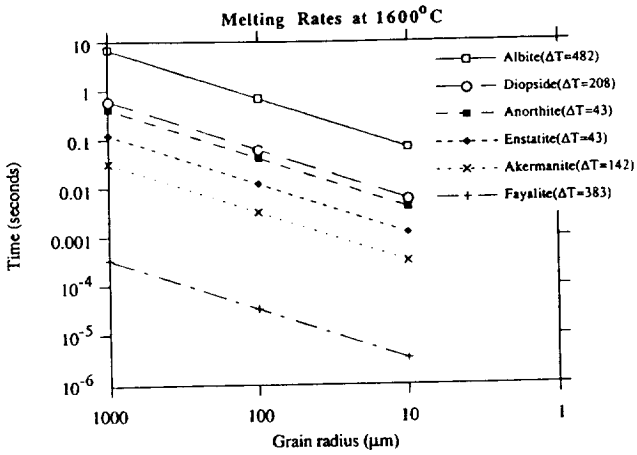

Fig. 2. Shown here is a time vs. grain radius plot for melting kinetics at 1600°C. For example, at 1600°C an enstatite grain with a radius of 1 mm will be completely melted in 0.12 seconds. (ΔT = superheat).

rates will necessarily increase rapidly (eqn. 5), suggesting that if peak temperatures were below 1901°C, the chondrule may consist of melt and relict forsterite as a result of high temperature heating. Heating above 1901°C will lead to complete melting of chondrules in seconds or less, provided that the minerals listed in Table 2 are a major proportion of the precursor assemblage.

Albite has been proposed as a chondrule precursor (Hewins, 1991a) to explain the 1:1 correspondence between Na and Al found in FeO-rich chondrules, yet relict albite is not found in chondrules. At 1600°C (Fig. 2), a 1 mm radius albite grain would be completely melted in less than 10 seconds. For grain radii in the 10 - 100 μm range, albite would be completely melted in less than 2 seconds at 1600°C. Using the experimentally determined uη vs. ΔT relationship shown in Fig. 1 for albite, the melting rates are 11μm/s and 144 μm/s at 1500°C and 1600°C, respectively. Clearly, if albite is fine-grained (1 – 10 μm) it could melt completely in a flash heating event with a peak temperature as low as 1500°C. If the peak temperature were higher (Hewins and Connolly, this volume) then it is not surprising that we do not find relict albite in chondrules.

Relict Grains

The presence of relict grains in chondrules (grains that survived the last heating event) can help constrain the maximum temperatures experienced by the host chondrules during heating. Enstatitic pyroxene and forsteritic olivine have been identified as relict grains in FeO-rich porphyritic chondrules (Jones, this volume). Dusty olivines have been identified as relict grains in FeO-poor porphyritic chondrules (Nagahara, 1981; Jones, this volume). The common existence of relict olivine in chondrules constrains the maximum temperature that the interiors of these chondrules experienced to the melting point of forsterite, 1901°C (Richet *et al.*, 1993). Chondrules containing relict enstatite could not have been heated in excess of 1577°C, the liquidus temperature of enstatite, for more than a few seconds. Enstatite melts incongruently, undergoing a peritectic reaction at 1557°C, forming a product of 95% liquid and 5% forsterite, by weight. At 1577°C enstatite melts congruently to a liquid of pure enstatite (Bowen and Anderson, 1914). We have studied the congruent melting of an incongruently melting compound above the liquidus in the plagioclase system (Ab_{89}), and find that the congruent melting model embodied in eqn. (5) is appropriate for this type of reaction (Greenwood and Hess, 1995). We therefore feel that the estimates for the melting rate of enstatite above 1577°C are valid (Table 2, Fig. 2). As mentioned earlier, incongruent melting is generally a slow process, rate limited by solid-state diffusion. The interested reader is referred to Tsuchiyama and Takahashi (1983) for a discussion.

DISCUSSION

The constraints developed in this paper are for minerals transforming to liquids via melting. The total destruction of a relict grain and/or precursor mineral involves the incorporation of this nominally pure mineral melt into the bulk chondrule melt. This is synonomous with liquid interdiffusion, and will necessarily take extra time. For example, enstatite melts at a rate of ~3700 μm/s, at 1600°C. This enstatitic liquid would then need to interdiffuse with the chondrule melt before cooling, otherwise it will recrystallize with an anomalous core composition.

The melting rates calculated above assume there is enough energy to melt chondrule precursor minerals, and that melting is not limited by the transfer of heat to or away from the interface. The observation that some chondrules were undoubtedly completely molten (e.g. Grossman, 1988) would seem to support this assumption. Heat-flow limited melting would occur if the interface could not maintain temperature, due to the loss of heat to the interior of the mineral or its surroundings. The low thermal conductivities of silicates (Kingery *et al.*, 1976) should prevent this from happening. The interested reader is referred to Spaepen and Turnbull (1982) for a review of heat-flow limited melting.

The loss of heat from chondrule mineral surfaces to the surroundings has recently been considered by Horanyi *et al.* (1995), wherein they modelled chondrule formation in lightning discharges. They conclude that there may not be enough energy available to completely melt silicates. In their paper, they model the loss of heat from mineral surfaces to the surroundings by radiative cooling, an assumption that is in direct conflict with constraints obtained from dynamic crystallization experiments (Hewins, 1988). Also, the loss of heat to chondrule surroundings would probably only apply to minerals near the exteriors of precursor aggregates, as minerals in the interiors would be thermally insulated by the newly formed molten chondrule. These factors suggest that melting of chondrule precursor mineral grains was not limited by heat flow.

CONCLUSIONS

1. Consideration of congruent melting kinetics explains why most chondrule precursors did not survive flash heating events. A congruent melting model can be used to calculate melting rates.

2. The presence of relict forsteritic olivine and enstatitic pyroxene in chondrules provides the source of constraints on the maximum temperature that the interiors of these chondrules experienced. Chondrules containing relict grains of forsterite and enstatite probably did not exceed temperatures of 1901°C and 1577°C, respectively, for more than a few seconds.
3. Albite can be a precursor and not a relict grain, even though it has sluggish melting kinetics. Specifically, the melting rates of albite at 1500°C and 1600°C are 11 μm/s and 144 μm/s, respectively, suggesting that fine grained albite (< 10 μm) would completely melt in seconds or less for a peak temperature as low as 1500°C.
4. Chondrule formation models that attempt to explain the melting event(s) need to take into account the kinetics of melting in order to constrain their formation characteristics.

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